SUMMARY OF THE INVENTION

Accordingly, an object of the present invention is to provide a dental restorative material comprising an ultrafine filler having a size of 0.1 μ m or less incorporated thereinto in a high density, which has high strength and high aesthetic appearance.

Another object of the present invention is to provide a dental retorative material with excellent durability, which can maintain high strength for a long period of time under wet conditions in the oral cavity where the material suffers repeated oclusal pressure.

The present inventors have investigated to solve the above-mentioned problems and found that the objects of providing high-density filling, high strength and high aesthetic appearance and improving durability can be achieved by the combined use of an ultrafine inorganic particulate filler with a size of 0.1 μ m or less surface-treated with a silane coupling agent comprising (a) a (meth)acryloyl group and a linear alkylene group having at least 8 carbon atoms and (b) a hydrophobic multifunctional (meth)acrylate; wherein the term "(meth)acryloyl group" herein means either methacryloyl group or acryloyl group, and the term "(meth)acrylate" means either methacrylate or acrylate.

Thus, the present invention provides dental restorative materials comprising:

(a) an ultrafine inorganic filler with a size of 0.1 μ m or less which is insoluble in water and surface-treated with a silane coupling agent represented by the following general formula (I) in an amount of at least 3% by weight based on the weight of the filler:

wherein R^1 is a hydrogen atom or a methyl group, R^2 is 40 a hydrolyzable group, R^3 is a hydrocarbon group having 1 to 6 carbon atoms, X is an oxygen or sulfur atom, m is 2 or 3 and n is an integer of 8 to 20; and

(b) a (meth)acrylate monomer composition containing at least 50% by weight based on the weight of the 45 composition of at least one hydrophobic multifunctional (meth)acrylate represented by the following general formula (II):

$$\begin{pmatrix}
R^{5} \\
H_{2}C = C - C - O \\
0
\end{pmatrix}_{p} R^{4}$$
55

wherein R⁴ is an organic group having 7 to 40 carbon atoms, being composed of 1 to 8 hydrocarbon groups having 2 to 40 carbon atoms, at least one of said hydrocarbon groups being a hydrocarbon group having at 60 least 4 carbon atoms, the ratio of the number of total carbon atoms, x, contained in said hydrocarbon groups to the number of the hydrocarbon groups, y, contained in said organic group satisfying:

x/y > 3; R⁵ is a hydrogen atom or a methyl group and 65 p is an integer of 2 to 8;

wherein said surface-treated inorganic filler is incorporated in an amount of at least 100 parts by weight based on 100 parts by weight of the monomer composition.

The most characteristic feature of the present invention resides in the use of an ultrafine inorganic filler having a particle size of $0.1~\mu m$ or less surface-treated with a silane coupling agent represented by general formula (I) in combination with a hydrophobic multifunctional (meth)acrylate represented by general formula (II). The term "particle size" herein means an arithmetic mean of the major axis and the minor axis of the particle.

In the hydrophobic multifunctional (meth)acrylate, interactions among the particles of the ultrafine inorganic filler, which particles have been made highly hydrophobic by surface treatment with the silane coupling agent of formula (I), are extremely weakened so that the thickening (viscosity increasing) effect of the particles decreases and the filler can be incorporated in a high ratio. Moreover, since the (meth)acrylate is multifunctional, a highly crosslinked three-dimensional network structure develops between the filler and the resin matrix, thereby exhibiting high strength.

In the silane coupling agent (I), examples of R² include chlorine atom, alkoxy group, isocyanate group, acyloxy group and iminoxy group, among which particularly preferred are chlorine atom, alkoxy group and isocyanate group because of their high activity. Examples of the silane coupling agent (I) are as shown below.

$$\begin{array}{c} \text{CH}_{3} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{8}} \, \text{Si} + \text{OCH}_{3})_{3} \\ \text{CH}_{3} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{11}} \, \text{Si} + \text{OCH}_{3})_{3} \\ \text{CH}_{3} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{11}} \, \text{Si} + \text{O} + \text{CH}_{2})_{\overline{5}} \, \text{CH}_{3}]_{3} \\ \text{CH}_{3} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{11}} \, \text{SiCl}_{2}, \\ \text{CH}_{3} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{11}} \, \text{SiCl}_{3} \\ \text{CH}_{3} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{11}} \, \text{Si} + \text{OCH}_{3})_{2} \\ \text{CH}_{3} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{11}} \, \text{Si} + \text{OCH}_{3})_{2} \\ \text{CH}_{3} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{11}} \, \text{Si} + \text{OCH}_{3})_{2} \\ \text{CH}_{3} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{11}} \, \text{Si} + \text{OCO})_{3} \\ \text{H} \\ \text{H}_{2}\text{C} = \text{C} - \text{COO} + \text{CH}_{2})_{\overline{11}} \, \text{Si} + \text{OCH}_{3} \\ \text{O} \\ \text{O$$

COO+CH₅)₁₃-Si+OOCCH₃)₃